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## **FNAS/LDEF RADIATION DATA ANALYSIS**

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## **ATMOSPHERIC RADIOACTIVE ISOTOPES AT ORBITAL ALTITUDES**

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### **Abstract**

The radioactive isotope  $^7\text{Be}$  was discovered on the forward-facing side of the LDEF satellite in amounts far exceeding that expected from direct cosmic ray activation of the spacecraft material. This prompted an examination of the production of cosmogenic isotopes in the atmosphere and of the processes by which they may be transported to orbital altitudes and adsorbed by a spacecraft.  $^7\text{Be}$  is only one of several atmospheric cosmogenic isotopes which might be detectable at orbital altitudes and which might prove to be as useful as tracers of atmospheric circulation processes in the mesosphere and thermosphere, as they have been in the lower layers of the atmosphere.

## 1. INTRODUCTION

The Long Duration Exposure Facility (LDEF) satellite was launched by the space shuttle *Challenger* on April 7, 1984 into a circular orbit of altitude 480km and inclination 28.5 deg. It was retrieved on January 12, 1990, 6 years later, by the *Columbia* from an orbit which had decayed to 310km due to atmospheric drag. The LDEF structure was a 12-sided cylinder 9.1m long by 4.3m in diameter. It was designed to maintain a stable attitude in orbit with its long axis vertical and the same one of its 12 sides always faced into the direction of travel when the satellite was not attached to the Shuttle. After orbital recovery, the satellite was subjected to extensive examination for radioactivity induced in the wide variety of materials carried on the satellite. These studies, along with measurements and calculations of the low Earth orbit radiation fields, were performed by a consortium lead by the NASA, Marshall Space Flight Center (MSFC), and have been extensively reported (Parnell, 1991; Harmon et al., 1996; Harmon et al., 1992; Armstrong et al., 1991, 1992; Watts et al., 1993).

## 2. THE DISCOVERY OF $^7\text{Be}$ ON THE LDEF SATELLITE

The first overall survey measurements of induced radioactivity on LDEF were made by the Naval Research Laboratory at the Kennedy Space Center immediately after return of the satellite and prior to the removal of any materials (Phillips et al., 1991a, b, c). The satellite was rotated about its long axis, rotisserie-style, next to an array of large germanium detectors. The  $^7\text{Be}$  activity measured on each of the 12 sides of the satellite is shown in Figure 1 (Fishman et al., 1991). The  $\gamma$ -ray line at 478 keV was unambiguously observed to emanate from the leading sides of the satellite, while the much weaker intensity from the trailing-edge sides represents flux from the leading edge attenuated by passing through the satellite body. Individual components were quantitatively measured in a low-level  $\gamma$ -counting facilities at MSFC and elsewhere (Harmon et al., 1992, 1996). Figure 2 shows the  $\gamma$ -ray spectrum from two identical anodized aluminum plates, one of which was exposed to the ambient atmosphere in orbit on the leading-edge of the vehicle and the other on the trailing-edge. The activity on the trailing-edge sample was two orders of magnitude less than that on the leading edge, and consistent with the computed level of  $^7\text{Be}$  produced in the body of the plate by spallation of Al atoms by cosmic ray protons during the orbital lifetime (Gregory et al., 1991). Figure 3 shows the activities of two isotopes,  $^7\text{Be}$  and  $^{22}\text{Na}$ , measured on individual aluminum clamp-plates from positions all around the spacecraft. The radically different distribution indicates different provenance. In general, radioactivities induced in materials on the trailing-edge, such as  $^{22}\text{Na}$  from the Al spallation, were higher than those in materials on the leading faces because of the anisotropy of the trapped proton flux which peaks from the south-west direction (Watts, Heckman and Parnell, 1989). Since the direction of the LDEF orbit was from west to east, these particles are more likely to interact with materials in the trailing-edge of the satellite. We were thus led to conclude that the  $^7\text{Be}$  must have accumulated on the leading surfaces by adsorption from the ambient atmosphere at orbital altitudes.

In Table 1, the measured number of  $^7\text{Be}$  atoms per unit area on various LDEF surfaces is shown (Fishman et al., 1991). The results are corrected to the retrieval date of 12 January 1990 and for the offset angle of the collecting plate from the leading direction. The areal density for  $^7\text{Be}$  on aluminum and steel is the same within the experimental uncertainty, and is apparently not a strong function of the type or surface condition of the metal. However, the polytetrafluorethylene (PTFE) thermal coating, which was used on many LDEF experiment trays, has a density of  $^7\text{Be}$ , an order of magnitude lower than that

found on the aluminum surface. The reason for this apparent difference in uptake efficiency is unknown, but could be related to the covalent-bond structure of the material. The explanation may be complicated also by the observed erosion of the PTFE surface by atomic oxygen.

**Table 1. <sup>7</sup>Be Concentrations on LDEF Leading Edge Surfaces of Various Chemical Composition.** Areal concentrations in atoms cm<sup>-2</sup> have been corrected back to the date of recovery, 12 January 1990, and for the angular offset of the collecting plate with respect to the direction of orbital motion.

Material	<sup>7</sup> Be Areal Density (x 10 <sup>5</sup> atoms/cm <sup>2</sup> )
Stainless steel trunnion face	5.3 ± 0.7
Polished aluminum plate-Exp. A0114	6.7 ± 1.0
Anodized aluminum experiment tray clamp	4.6 ± 0.5
Teflon thermal cover	0.9 ± 0.2

From the measured densities of <sup>7</sup>Be on LDEF surfaces and making some simplifying assumptions, we can estimate the concentration of <sup>7</sup>Be atoms per cm<sup>3</sup> of air at the LDEF orbital altitude. Since the lifetime of LDEF is much greater than the mean lifetime of a <sup>7</sup>Be atom, and ignoring changes in altitude over the last 6 months in orbit, we assume a steady state relationship between pickup of <sup>7</sup>Be and loss by decay:

$$\frac{dn}{dt} = 0 = -k n_{eq} + n^* v p_s$$

where:  $n$  is the density of <sup>7</sup>Be atoms on the surface at time  $t$   
 $k$  is the first-order decay constant for <sup>7</sup>Be  
 $n_{eq}$  is the steady-state surface density of <sup>7</sup>Be in atoms cm<sup>-2</sup>  
 $n^*$  is the concentration of <sup>7</sup>Be atoms in orbital space (atoms cm<sup>-3</sup>)  
 $v$  is the spacecraft velocity (cm s<sup>-1</sup>)  
 $p_s$  is the sticking probability of Be on a metal surface

$k = \frac{\ln 2}{t_{1/2}}$ , where:  $t_{1/2}$  is the half life, 53.2d. Thus we have:  $n_{eq} = n^* v t_{mean} p_s$ , where  $t_{mean} = \frac{t_{1/2}}{\ln 2} = 76.8$  days for <sup>7</sup>Be. From the measured value of  $n_{eq}$ , assuming  $p_s = 1$ , we have  $n^* = 1.2 \times 10^{-7}$  cm<sup>-3</sup> at 320 km or a relative concentration of  $3.8 \times 10^6$  atoms per gram of air. In the peak production region, below 20km, previous measurements (Benioff, 1956; Lal et al., 1958; Lal and Peters, 1967; Bhandari, 1970; O'Brien, 1979) yield a concentration of 1000 <sup>7</sup>Be atoms per gram of air, in agreement with a simple calculation using known values of the cosmic-ray flux and the production cross-section for the isotope. Thus, the measured concentration of <sup>7</sup>Be per unit mass of air at 320km is three to four orders of magnitude greater than it would be if it had been produced by cosmic ray spallation at that altitude.

The simplest explanation is that Be is quickly transported upwards from regions of the atmosphere where its numerical concentration is much higher (but not its relative concentration with respect to oxygen and nitrogen). This transport must take place on time scales similar to or shorter than the radioactive half-life (53.2 days).

## 2.1 Production and Transport of Isotopes in the Atmosphere

Before an isotope may be used as a tracer for geophysical processes, the production and removal rates must be known both in space and time. These are often called the source- and sink-functions and are considered to be reasonably well understood for Be from theoretical and experimental standpoints.

The Earth and its atmosphere are continuously bombarded by particulate radiation including galactic cosmic rays and particles from the Sun. Primary nuclei of energy more than a few 10's of MeV per nucleon may cause nuclear transformations of matter, changing its elemental and isotopic composition. Most of these transformations at Earth occur in the atmosphere, with the primary production zone occurring in the stratosphere and upper troposphere (Lal and Peters, 1967). Almost all of the production is by nucleons, with a very small component being produced by secondary mesons, muons and  $\gamma$ -rays. The latter component is much more important at or below ground level, but does not concern us here.

Nuclei produced by spallation of atmospheric atoms include a large number of stable isotopes. These cannot usually be identified by source, due to the presence of large quantities of ordinary matter with which the cosmogenic material becomes rapidly mixed. This is not the case with some stable cosmogenic nuclides in meteorites. New technologies may allow the fixing of stable atmospheric cosmogenic isotopes onto surfaces before contamination with ordinary material occurs and permit identification for example by surface ion mass spectroscopy (SIMS). To the present time, radioactive cosmogenic isotopes have been identified and used as tracers for a variety of geophysical processes including circulation in the atmosphere and the oceans, sedimentation, ice accumulation and many other applications (see Lal and Peters, 1962, 1967).

The most abundant isotopes produced in the atmosphere are those derived from N and O and are therefore of light mass and few in number. Radioisotopes derived from Ar and Kr are present but at much lower levels and are not discussed further here. A list of radioactive isotopes formed by spallation of N and O and identified in rainwater is shown in Table 2.

**Table 2. Cosmogenic radioisotopes produced in the atmosphere from nitrogen and oxygen target nuclei**

Isotope	Half-life	Radiation
$^{10}\text{Be}$	$2.5 \times 10^6\text{y}$	$\beta$ , 550 keV
$^{14}\text{C}$	$5.73 \times 10^3\text{y}$	$\beta$ , 156 keV
$^3\text{H}$	12.3y	$\beta$ , 18 keV
$^7\text{Be}$	53d	$\gamma$ , 480 keV

Beryllium has 7 isotopes, of which only one,  $^9\text{Be}$ , is stable. Two isotopes have half-lives long enough to be of use in atmospheric and geophysical tracer work. These are  $^7\text{Be}$ ,  $t_{1/2}=53.2$  d and  $^{10}\text{Be}$ ,  $t_{1/2} = 1.5 \times 10^6$  y.  $^7\text{Be}$  decays by electron-capture and 478 keV  $\gamma$ -emission to  $^7\text{Li}$ . This  $\gamma$ -ray line emission may be measured at very low levels (a few 100's of counts per day) by large Ge detectors in low-background counting facilities.  $^{10}\text{Be}$  decays by internal conversion to  $^{10}\text{B}$  emitting a fast electron with a wide spectrum of energies. Discrimination of this radiation from natural background is much more difficult except at high activities. Fortunately, the method of accelerator mass spectrometry (AMS) is now available, which permits measurements of concentrations of  $^{10}\text{Be}$  (as well as  $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$  and others) at activities much lower than generally possible by radioactivity counting methods. The chief advantage of the method is that one measures the actual number of  $^{10}\text{Be}$  atoms in the sample, not the very small fraction of those which decay during the measurement time. This is particularly important where the sample available is very small, such as for  $^{14}\text{C}$  measurements in valuable or rare historical materials, or in our case, for the  $^{10}\text{Be}$  samples from satellite surfaces, which are just intrinsically small. A disadvantage of the method in many cases is the need for elaborate chemical preparation of the sample prior to insertion into the ion-source of the mass spectrometer.

$^7\text{Be}$  was first detected in Indian monsoon rainwater (Goel et al., 1956) and in Chicago rainwater (Arnold and Al-Salih, 1955).  $^{10}\text{Be}$  was discovered in deep-sea sediments (Arnold, 1956). Since that time, there have been several studies which have demonstrated the usefulness of  $^7\text{Be}$  as a tracer for stratosphere/troposphere exchange (Viezee and Singh, 1980).  $^{10}\text{Be}$  has been used extensively as a tracer in geology, glaciology and oceanography. Deposition to the Earth's surface of both isotopes increases during (or immediately following) seasonal weather disturbances of the stability of the tropopause. The ratio of  $^{10}\text{Be}$  to  $^7\text{Be}$  has been used to measure the residence times of these isotopes in the stratosphere where about 70% of the atmospheric production occurs (Raisbeck et al., 1981). The ratio of production rates,  $^{10}\text{Be}/^7\text{Be}$ , is approximately 0.5 (O'Brien, 1979) while the half-lives are very different, that of  $^{10}\text{Be}$  being much longer than the loss-rate from the atmosphere. Measurements of steady-state concentrations of the two isotopes in the stratosphere showed ratios of  $^{10}\text{Be}/^7\text{Be} = 2.6$  to 3, implying an average residence time of 320 to 350 days (Raisbeck et al., 1981). This characteristic mixing time may not hold at higher altitudes in the stratosphere and mesosphere, where longer residence times might yield a larger  $^{10}\text{Be}/^7\text{Be}$  ratio. This would have implications for a proposed model for the distribution profiles of the two isotopes suggested by Petty (1991).

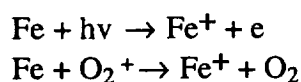
The removal process is considered to be reasonably well understood in the troposphere and lower stratosphere (Figure 4). Data at higher altitudes, particularly those of interest here (above 100 km) are non-existent prior to LDEF. Isotopes created at 12-20 km which constitute the bulk of production are rapidly oxidized to normal oxides,  $^7,^{10}\text{BeO}$ ,  $^{14}\text{CO}$  and  $^{14}\text{CO}_2$ , and  $\text{H}_2\text{O}$  (containing one  $^3\text{H}$  atom). Sink functions for these oxides differ considerably due to their very different chemistry. Be oxides attach themselves rapidly to sulfuric acid and solid aerosols of sub-micron size which are widely dispersed in the lower stratosphere (Junge, 1963), and which are small enough that they move convectionally with the local atmosphere with negligible gravitational settling. In the troposphere these aerosols are rapidly removed by water-droplet formation and rain fallout. Residence time in the troposphere is short, about 1 month (Lal and Peters, 1962), due to the efficiency of the scavenging process. The Be isotopes are then transported to the land and oceans by rainfall. Turbulent mixing, or convection, across the tropopause is slower,

with a mean residence time in the stratosphere for aerosols and attached isotopes of about one year, at least in the lower stratosphere.

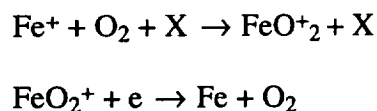
## 2.2 Atmospheric Chemistry of Be

As very little information is available on the chemistry of Be neutrals and ions in low density gases, we have reviewed the chemistry of some metallic analogs observed by meteor ion chemistry. Metal (Mg, Ca, Al, Fe) and silicon atoms and ions are deposited in the lower thermosphere by meteor ablation. The chemistry of these species has been reviewed by Solomon et al., 1982.

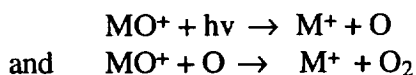
The form of the meteoritic ions is highly variable with altitude and between day and night. Electropositive metals readily form positive ions:



At low altitudes neutralization may occur (X is a third molecule):



In general, at altitudes in excess of 100km, the metal (M) oxides cannot survive in appreciable quantities due to reactions such as

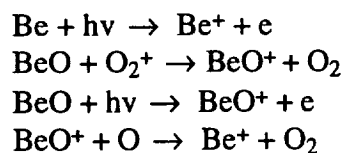


Thus at higher altitudes, the singly-charged positive ion dominates for most metallic species studied. Of interest is the ratio  $\text{M}^+/\text{M}$  which varies with altitude and electropositive character of the metal. Examples of some measured ratios from the literature (Solomon et al., 1982) for silicon and iron are:

$$\begin{aligned}\text{Si}^+/\text{Si} &= \begin{array}{l} 0.006 \text{ at } 96\text{km} \\ 0.2 \text{ at } 110\text{km} \end{array} \\ \text{Fe}^+/\text{Fe} &= \begin{array}{l} 5 \text{ at } 100\text{km} \\ 220 \text{ at } 110\text{km} \end{array}\end{aligned}$$

Thus above 150km (and perhaps as low as 100km) most Be should exist as  $\text{Be}^+$ .

Important reactions might be:



Closer to the turbopause, in the region 90 to 110km altitude, a proposed source region for the orbital Be, Si chemistry shows some differences from that of the metals. Ferguson et al. (1981) suggest the formation of an appreciable fraction of  $\text{SiOH}^+$ . A question remains as to whether the chemistry of Be is more similar to that of the metals or of Si. Persistence of BeO species at 100km would affect vertical diffusion models due to the larger mass.

### 2.3 The Adsorption Process of Be on Satellite Surfaces

To determine absolute concentrations of atmospheric species by the satellite surface reaction or uptake technique, the reaction efficiency or the sticking probability between the molecule and the surface materials must be known.

Polymeric films of organic materials such as the polyimide, Kapton and polytetrafluoroethylene (PTFE), which are often employed on exterior surfaces of satellites make poor adsorbers for the metallic ions discussed here. Firstly, bonding may not readily occur between the covalent polymer and a metal ion such as  $\text{Be}^+$  or an ion-pair such as BeO. Secondly, most organic polymers, including the two above, are oxidized and eroded by the flux of oxygen atoms present in the orbital atmosphere which impact the front surfaces of satellites at orbital velocity (about  $8 \text{ km s}^{-1}$ ) just as the Be ions do. The lower  $^7\text{Be}$  concentration measured on PTFE (Table 1) is probably due to one or both of these causes. Whatever the chemistry of Be is at 100km, it seems clear that at satellite altitudes, the bulk of the metals are in the form of singly charged monatomic species, in this case,  $\text{Be}^+$ . Since virtually all metal surfaces are covered with a layer of normal oxide under these conditions (Gregory et al., 1986), a model is proposed in which a fast  $\text{Be}^+$  ion enters the surface metal oxide lattice where it is rapidly thermally accommodated and trapped. The flux of atomic oxygen also present under these conditions will satisfy the needs of stoichiometry.

We investigated the form of binding of the Be to the aluminum surfaces on LDEF (Fishman et al., 1991). Possibilities included (1) binding within an adsorbed contaminant layer, for example of hydrocarbon; (2) binding of Be-containing particulates, perhaps aerosols or meteoritic debris and (3) binding within the native oxide found on aluminum and other metals. Two kinds of Al plates from the LDEF were measured, some with several microns of oxide produced by anodization and the second type a polished Al plate from the UAH Atomic Oxygen Experiment A0114. The oxide on this was only expected to be 50-100 Å thick.

The polished Al plate was coated with a solution of Collodion, which was then dried, stripped off and counted. No Be activity could be associated with the Collodion film. The method is used in industry to reliably and quantitatively remove particulates from sensitive surfaces. Next the plate was wiped first with alcohol, then with xylene. No activity was removed with the wipes. Finally an acid etch was used to remove the top 10 microns of the surface. The etch solution contained most of the Be activity formerly on the plate; that remaining being associated with either unetched surface or with re-adsorption of  $\text{Be}^{2+}$  ions onto the Al. This latter might be expected since a stable Be carrier solution was not used. The experimental results are consistent with the hypothesis that the Be species were penetrating the aluminum oxide layer on the surface of the plates and becoming permanently fixed in the oxide lattice. We believe the penetration to be of the order of one nanometer, since the kinetic energy of the Be species relative to the spacecraft was only 2.5eV. We did not have the capability to remove such a thin layer from large areas of metal surfaces, and thus could not measure a depth/composition profile for the species.



The data (Table 1) on Al and stainless steel substrates on LDEF (Fishman et al., 1991) shows similar concentrations of  $^7\text{Be}$  within experimental error. While this does not prove that the sticking probability,  $p_s$ , of  $^7\text{Be}$  on both equals unity, it is a reasonable deduction. Values of  $p_s < 1$  would of course yield even higher atmospheric concentrations of  $^7\text{Be}$  at orbital altitudes. We note that measurements of the  $^{10}\text{Be}/^7\text{Be}$  ratio (when these are achieved) should be insensitive to the value of  $p_s$ .

## 2.4 Tracers of Motion in the Upper Atmosphere

In this section we explore the use of satellite surfaces for trapping atmospheric radioisotopes and some possibilities for using such data as a tracer for atmospheric flows and models.

In principle, cosmogenic radioactive tracers should prove as useful a tracer of atmospheric flows and residence times in the upper atmosphere as they have in the troposphere and stratosphere. Some differences are apparent which affect both the source and sink functions. These arise because of (1) the transition from turbulent to molecular flow which occurs in the region 80-110km, and (2) different chemistry because of the more intense radiation, higher temperatures, and low gas density and (3) the increasing ratio of positive ions to neutrals for metal species above 90 km.

Petty (1991) has described a simple model in which Be isotopes produced in the uppermost layer of the well-mixed homosphere (nominally at 100km; see Figure 4) achieve gravitational equilibrium in a short period of time compared to their average lifetime, by diffusing upwards in an essentially free molecular flow regime.

Petty's model predicts a  $^7\text{Be}$  concentration of  $2.9 \times 10^{-8}$  atoms  $\text{cm}^{-3}$  at 310km for the active Sun case (asymptotic thermospheric temperature of 2000K). This is within a factor of 4 of the value deduced by Fishman et al. (1991), and by lowering the nominal altitude of the turbopause from 100km to 90km, where the source region is 5 times as dense, closer agreement can be obtained. The model makes clear that  $^7\text{Be}$  production in the upper mesosphere is adequate and reasonable to supply the enrichment factors observed above 300km and that transport from high  $^7\text{Be}$  production regions in the lower stratosphere is not necessary. However, as Petty points out, the model is based on the premise that the bulk of the Be within the domain of the model is in the form of free, neutral atoms. There is ample evidence that this is unlikely to be the case.

It is reasonable from the above discussion that the orbital measurement of  $^7\text{Be}$  (and  $^{10}\text{Be}$ , when that is successfully made) is sampling a production zone in the upper mesosphere. In the case of a diffusion model, the light isotopes distribute themselves according to their scale height in the atmosphere. A measurement of the  $^{10}\text{Be}/^7\text{Be}$  ratio, after correction for the scale height difference, would provide an estimate of the isotope ratio in the mesosphere, and thus of the characteristic residence time, or mixing time of the mesosphere. The current understanding of the  $^{10}\text{Be}$  cycle in the Earth's surface layers is shown in Figure 5.

There are some questions on which inadequate information exists to define a transport model.

1. The diffusion model requires the Be to be present as free neutral atoms. Above 150km, and perhaps even above 90km, the Be-containing species are likely to be positive ions. At this point, we do not know if transport of  $\text{Be}^+$  would be enhanced, inhibited or unaffected on average compared to neutrals. Spiro et al. (1979) have invoked vertical upward ion drifts to explain some observations. Such a mechanism could efficiently transport  $\text{BeO}^+$  and  $\text{Be}^+$  and would be independent of gravitational effects. In such a case, the  $^{10}\text{Be}/^7\text{Be}$  ratio would reflect the source region directly without correction for the different scale heights of the two isotopes.
2. The success of the diffusion model depends critically on the altitude of the turbopause to achieve the required source concentration of Be isotopes which then migrate upwards by diffusion to achieve the enrichment factor observed at 320km. The pressures at 80 to 100km indicate a transition from viscous to free molecular flow and a time-dependent calculation is needed to see if the process is fast enough to prevent significant loss of  $^7\text{Be}$ .
3. The proportion of Be in the form of oxide at 90-100km is unknown at this stage, but if significant, this species would be too massive to populate the thermosphere by diffusion.

### 3. CONCLUSIONS

The presence at orbital altitudes of the atmospheric cosmogenic nuclide  $^7\text{Be}$  was observed for the first time by its discovery on the LDEF spacecraft. It appears unlikely, however, that this aggregation could have any significant effects on the performance of satellite instruments or systems.

More measurements of  $^7\text{Be}$ , and new measurements of  $^{10}\text{Be}$  densities at orbital altitudes, are needed to refine understanding of the upward motion of these cosmogenic isotopes from the top of the mesosphere. Measurements of the  $^{10}\text{Be}/^7\text{Be}$  ratio will allow a better test of the diffusion model and investigate the possibility of other mechanisms associated with ion flows. The ratio will also allow determination of the characteristic mixing time of the upper mesosphere or turbosphere. At this time, the effects of large solar flares cannot be discounted as such an event occurred some weeks before recovery of the LDEF.

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## APPENDIX: THE SEARCH FOR $^{10}\text{Be}$ ON SATELLITE SURFACES

As discussed above, after  $^7\text{Be}$ ,  $^{10}\text{Be}$  is considered to be the most likely cosmogenic atmospheric radioisotope to be found on spacecraft surfaces. However, to our knowledge, it has not yet been identified on spacecraft from that source. A search was conducted on recovered LDEF surfaces, as described below, with interesting results which did not however contribute to the understanding of radioisotopes in the upper atmosphere.

We should expect to find the two Be isotopes together because of identical chemistry and similar source functions.  $^{14}\text{C}$  is probably present in the mesosphere as  $\text{CO}$  and  $\text{CO}_2$ . Molecules of these masses should not be significantly present at orbital altitudes, but even if present, they would not be expected to adhere to metal/metal oxide surfaces which mostly comprise outer spacecraft surfaces. Other well-known tracers such as  $^{26}\text{Al}$ , a product of Ar spallation, were not expected to be found. The production rate of  $^{26}\text{Al}$  in the atmosphere is only about  $10^{-3}$  that of the Be isotopes, and its mass 3 times larger.

$^{10}\text{Be}$  is measured at very low concentrations ( $10^5$  -  $10^6$  atoms per g. matrix) by accelerator mass spectrometry (AMS). Samples of material flown on the front surface of LDEF were prepared by etching the surface (in the case of Al tray clamps) or by machining a thin layer off and dissolving the turnings, in the case of the single stainless-steel sample (a trunnion-end). In each case it was necessary to take up about 1g of the metal sample into solution. The extractive and preparative chemistry was performed at Rutgers University (Albrecht et al., 1992) and the AMS analysis at the University of Pennsylvania (Middleton and Klein, 1987). The samples, after processing to remove Al, Fe, etc. and the addition of 1 mg of Be carrier (known to have very low  $^{10}\text{Be}$  content) were finally precipitated and transferred as solid BeO for AMS analysis. Samples from LDEF were restricted in their size, chemical composition, and availability. Most of the area on the front surface of the satellite was anodized Al except for significant amounts of poly-tetrafluorethylene (PTFE) and other thermal-control cratings. The PTFE thermal blankets showed low  $^7\text{Be}$  counts and were not analyzed for  $^{10}\text{Be}$ . Other non-aluminum metal parts such as screw heads and grounding straps were either of too small an area to yield a significant  $^{10}\text{Be}$  count by AMS, or were not available for analysis.

The first AMS results on front-facing Al clamp plates showed significant  $^{10}\text{Be}$  quantities. However, analysis of rear-facing and unexposed controls revealed  $^{10}\text{Be}$  where none was expected, including on clamp plates which had never left the Earth. These results suggested that  $^{10}\text{Be}$  might be present in significant quantities in commercial Al and its compounds, and a check of some of these materials was undertaken. The results (Middleton et al, 1994) are shown in Table 3. Details of the analysis and implications are given in that paper and only salient points summarized here.

### $^{10}\text{Be}$ in Aluminum and Bauxite Samples

The  $^{10}\text{Be}$  concentrations measured in various samples of Al metal, including LDEF tray clamps and household Al foil, varied from 40 to  $110 \times 10^6$  (atoms  $^{10}\text{Be}$ )/g Al, while concentrations in bauxite ore varied more widely, from  $15 \times 10^6$  atoms/g Al to  $6 \times 10^9$  atoms/g Al in different ore bodies. While cosmic rays at the Earth's surface do produce  $^{10}\text{Be}$  in soils, the production rate is very low compared to that in the stratosphere and yields

maximum steady-state concentrations of about  $12 \times 10^6$  atoms  $^{10}\text{Be}/\text{g SiO}_2$ . As all the Al samples and in particular the Haitian bauxite sample showed much higher concentrations than this, it is clear that most of the  $^{10}\text{Be}$  in these ores is not being produced *in situ*. As bauxite is an ore produced by surface weathering of older rocks, it is perhaps not surprising that it can easily be contaminated by  $^{10}\text{Be}$  produced in the atmosphere and carried to the surface in rainwater. As ore bodies vary greatly in their history of exposure to the atmosphere and subsequent burial under other soils, a wide variability of  $^{10}\text{Be}$  concentration may be expected. Middleton et al. show by some simple calculations that the amounts of  $^{10}\text{Be}$  found in bauxite are reasonably well accounted for by this mechanism. For example, if all the  $^{10}\text{Be}$  produced in a time similar to its 1.5 million year half-life washed out of the atmosphere (assuming even production and washout across the Earth's surface) were retained in a column of ore 1m deep, then the concentration of  $^{10}\text{Be}$  would be  $10 \times 10^9$  atoms/g of ore. This maximum concentration is similar to that seen in the Haitian bauxite. On the other hand, if the ore body were only exposed for 100,000y, but the  $^{10}\text{Be}$  carried were distributed over a 20m column, the average  $^{10}\text{Be}$  concentration would be about  $20 \times 10^6$  atoms/g ore.

The observation that commercial Al contains  $\sim 10^8(\text{atom } ^{10}\text{Be})/\text{g Al}$  has some practical consequences for users of AMS. Sometimes  $^{10}\text{Be}$  and  $^{26}\text{Al}$  assays are performed on the same samples necessitating use of both stable Be and Al carriers. If the Al carrier is prepared from material derived from a recently exposed ore body, it may contain significant quantities of  $^{10}\text{Be}$ . Similarly, Al sample holders should not be used in the AMS ion-source when  $^{10}\text{Be}$  assays are being performed. It was noted that Al ore bodies typically contain 1-10 ppm of normal  $^9\text{Be}$ . On average, the refining processes associated with production of the metal Al result in a lowering of the Be/Al ratio by an average factor of about 70. This would apply to  $^{10}\text{Be}$  as well as  $^9\text{Be}$ . The analytical grade  $\text{Al}_2\text{O}_3$  which was also analyzed for  $^{10}\text{Be}$  (Table 3) showed a lower ratio of  $^{10}\text{Be}/\text{g Al}$  than typical metal samples. This may reflect the effects of further purification steps which had been performed on this material.

#### $^{10}\text{Be}$ on Stainless Steel

Table 3 also includes an assay of a piece of commercial stainless steel from the laboratory.  $^{10}\text{Be}$  content was very low and we have proposed this material as a candidate substrate for further Be investigation on satellites. The single assay of satellite surface material, the circular end of a stainless steel trunnion from the LDEF, yielded an inconclusive result. No new results on cosmogenic isotopes at orbital altitudes have been obtained to our knowledge since the return of LDEF. However, an experiment is underway (Phillips, 1995) at present to expose a stainless steel (type 304) foil on a Russian Cosmos-series satellite. This will be subjected to both  $^7\text{Be}$  and  $^{10}\text{Be}$  assays after return from orbit.

## FIGURE CAPTIONS

Fig. 1 The activity of  $^7\text{Be}$  measured from the twelve sides of the LDEF spacecraft at the Kennedy Space Center before disassembly of components (from Fishman et al., 1991).

Fig. 2 Gamma-ray spectra of aluminum clamp-plates on the leading (a) side and trailing (b) side of the LDEF measured in the laboratory.  $^7\text{Be}$  is identified clearly on the leading side only, by its line at 478 keV. The strong line at 511 keV represents positron-annihilation gamma-rays from  $^{22}\text{Na}$  induced by spallation of Al nuclei, as well as some laboratory background. Both spectra were 48h counts (Fishman et al., 1991).

Fig. 3  $^7\text{Be}$  and  $^{22}\text{Na}$  activities for aluminum tray clamps taken from all round the LDEF. The leading edge is 0 deg. and the trailing edge 180 deg.  $^{22}\text{Na}$  is present in material from all sides, with a 2:1 predominance of the trailing:leading edges, while  $^7\text{Be}$  is essentially only present on the leading edge (from Gregory et al., 1991).

Fig. 4 Atmospheric structure, showing the nomenclature of the various layers and transitions discussed in the text. The layers of the ionosphere, beginning at about 85km are not included, (from Brasseur and Solomon, 1986).

Fig. 5 Cycle of cosmogenic  $^{10}\text{Be}$  in the Earth's surface layers (adapted from McHargue and Damon, 1991). Inventories are given in moles per year (M/y) and residence times as  $\tau$ .



Appendix. Table 3  $^{10}\text{Be}$  concentrations of aluminous materials (from Middleton et al., 1994)

Sample	Source	Mass [mg]	MassAl <sup>1</sup> [mg]	$^{10}\text{Be}/^9\text{Be}$ [10 <sup>-15</sup> ]	$^{10}\text{Be}$ [10 <sup>6</sup> atom/g Al]	Be [10 <sup>-9</sup> g/g Al]
Al AA Sol'n		140.0	140.0	63±4	61±3	159
Al foil		327.3	327.3	102±7	41±3	58
Al plate	LDEF2	256.7	256.7	122±8	75±3	
	LDEF 9-73	219.7	219.7	101±6	63±5	
	Shop	315.2	315.2	223±22	111±11	140
Bauxite	NBS 69A	1371.8	381.3	74±10	22±2	
Bauxite, Ark.	A 21485 <sup>4</sup>	361	141.5	48±4	15±2	
Bauxite, Haiti		497.7	52.2	33000±200	57200±3800	
Blank <sup>5</sup>				6±2		
Blank <sup>6</sup>		2085		5±1		

Notes: 1) Aluminum concentrations in the bauxites from Arkansas and Haiti were determined by DCP analysis to be 39.2% and 10.5%, respectively; NIST bauxite NBS 69A was used as a standard (27.8% Al). 2) Not flown. 3) Flown. 4) Label given by the American Museum of Natural History (New York, NY, USA). 5) Reagent blank. 6) Procedural blank run with stainless steel.

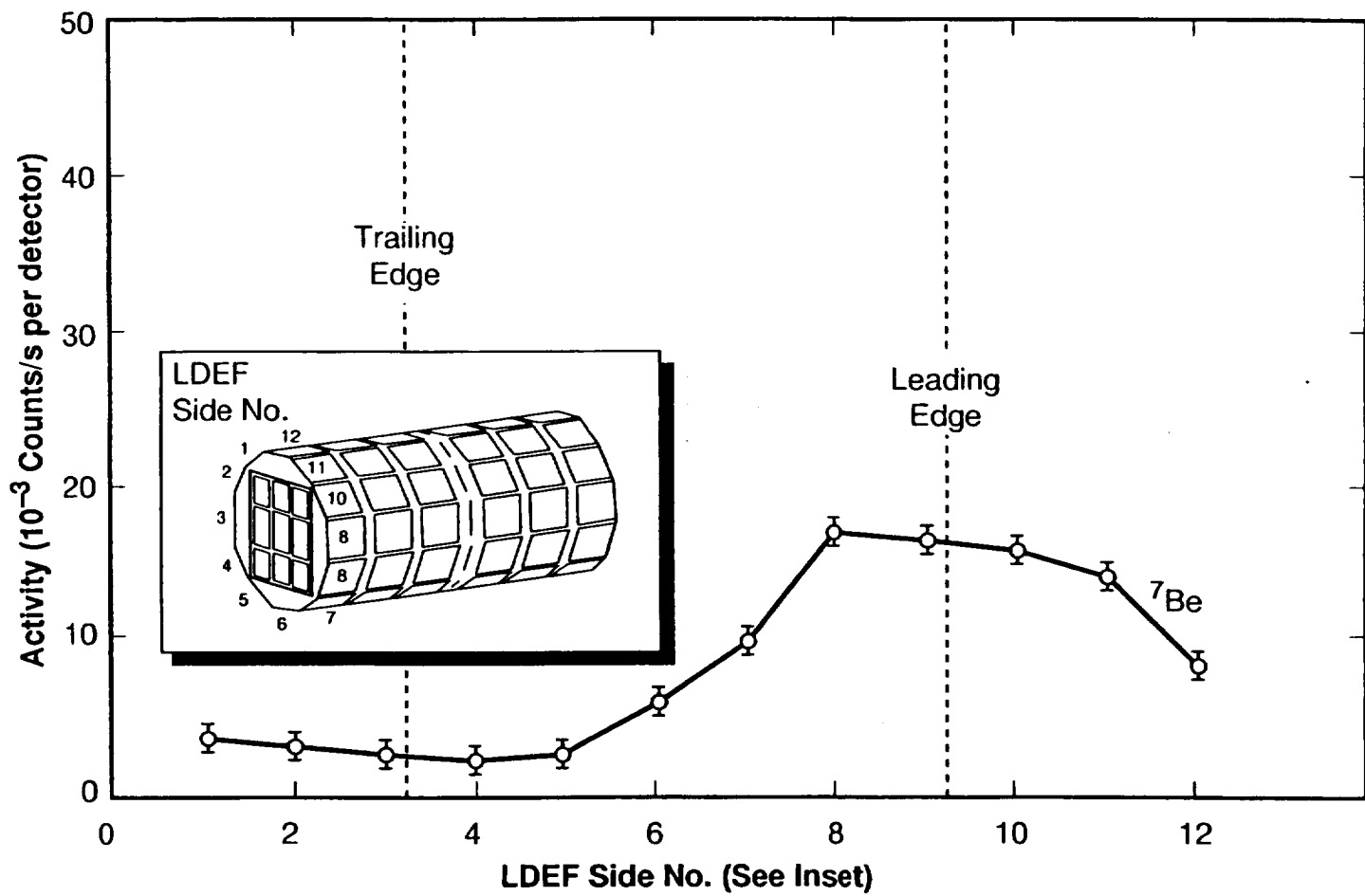
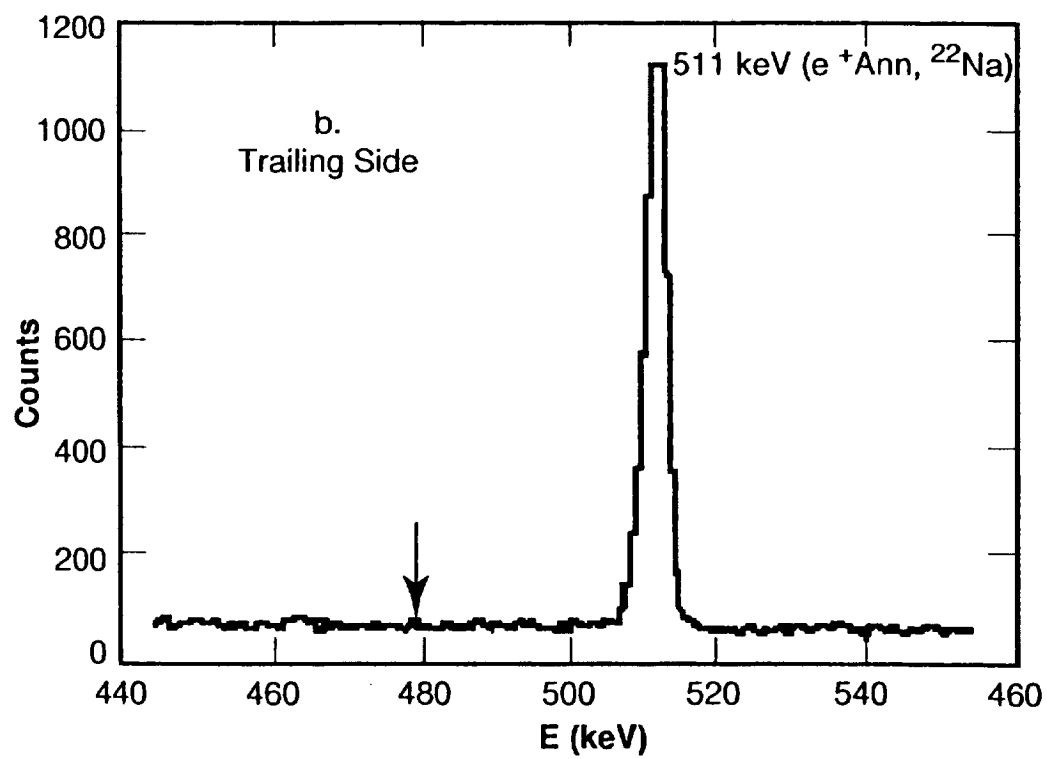
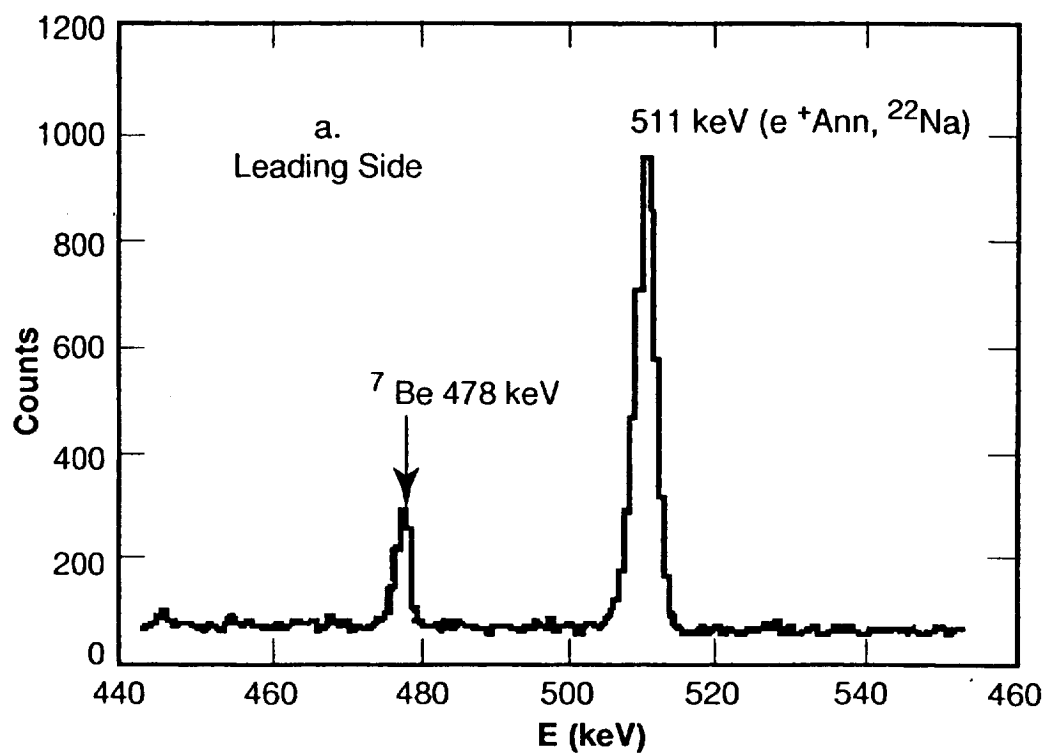


Fig 1



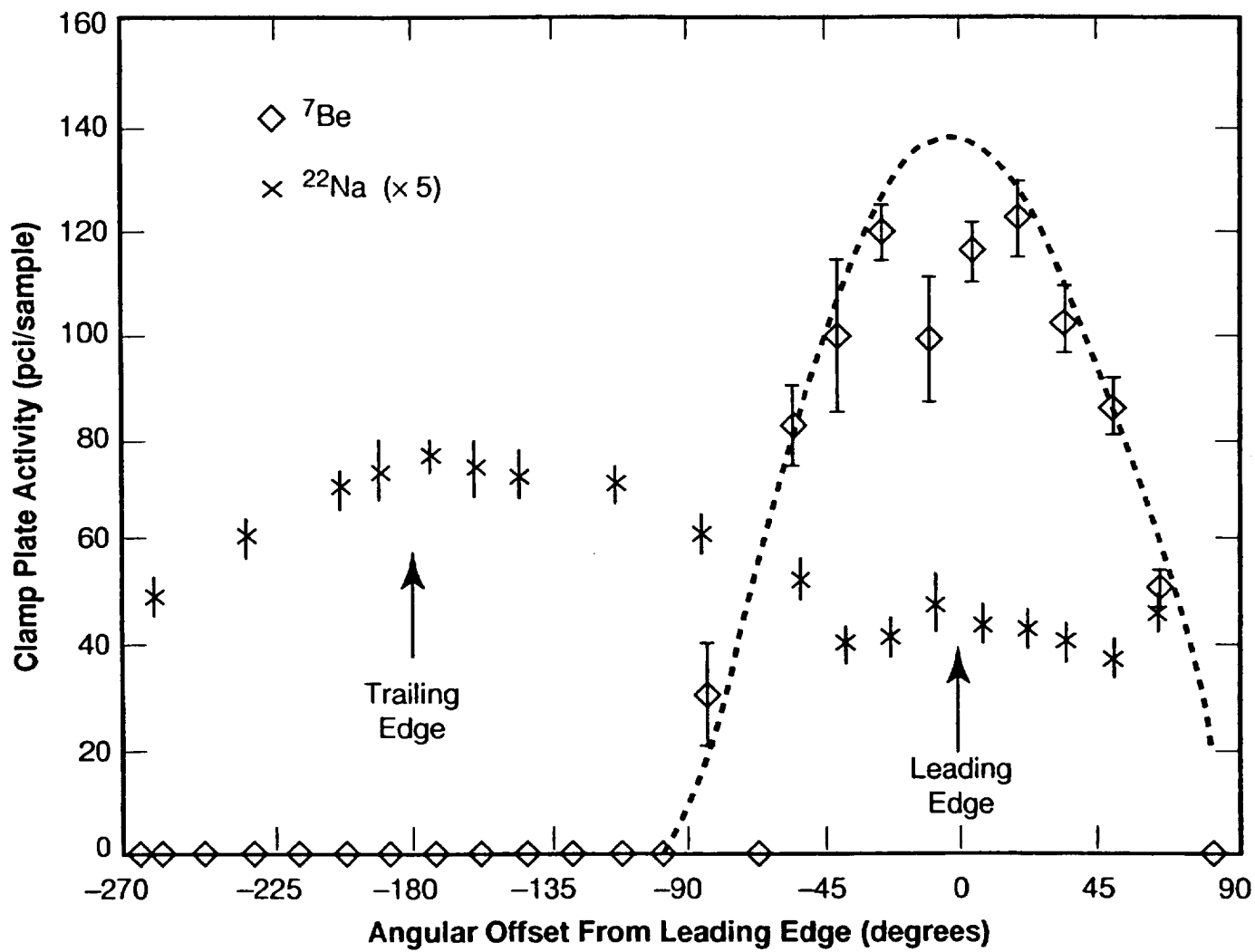
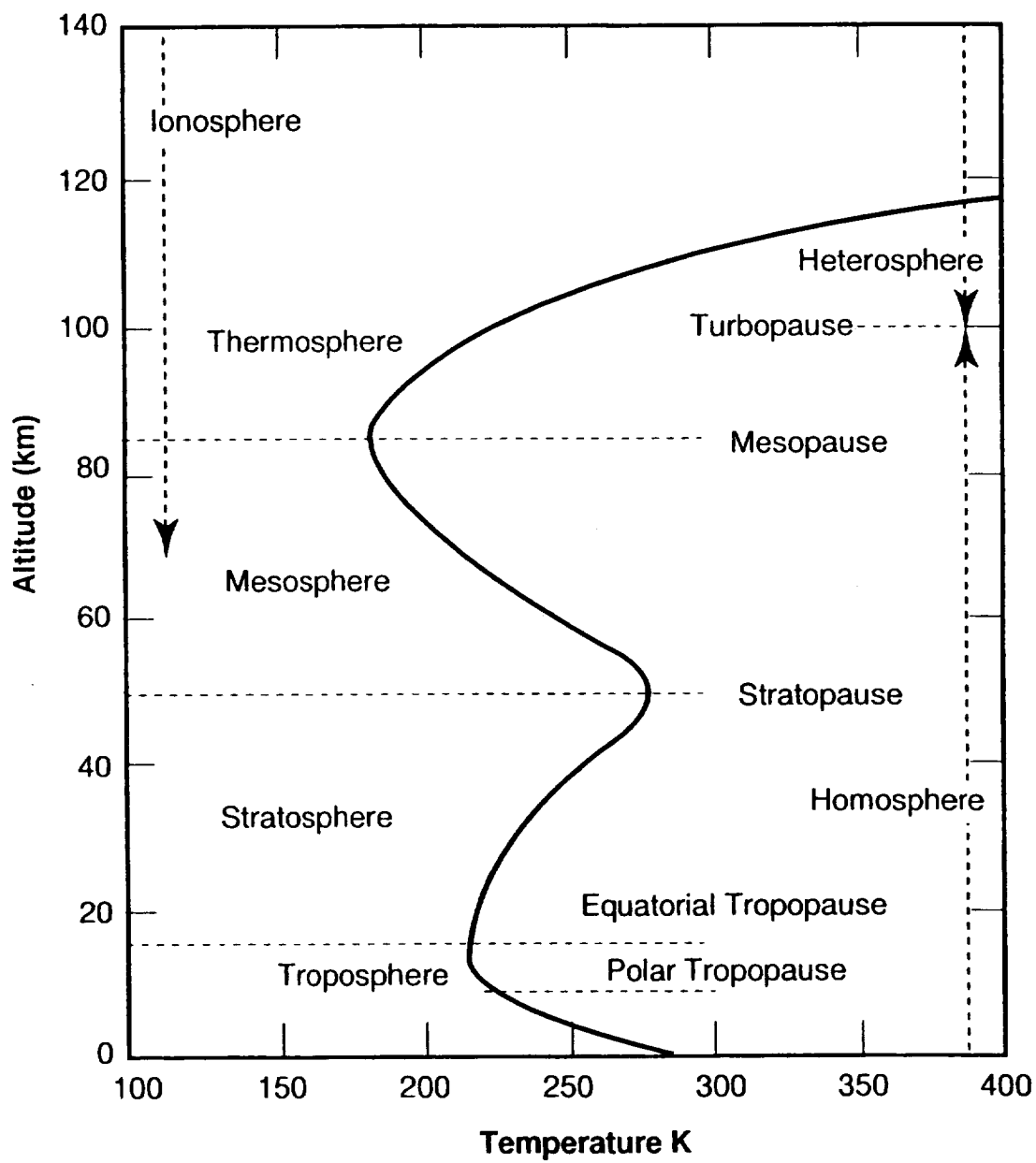


Fig 3



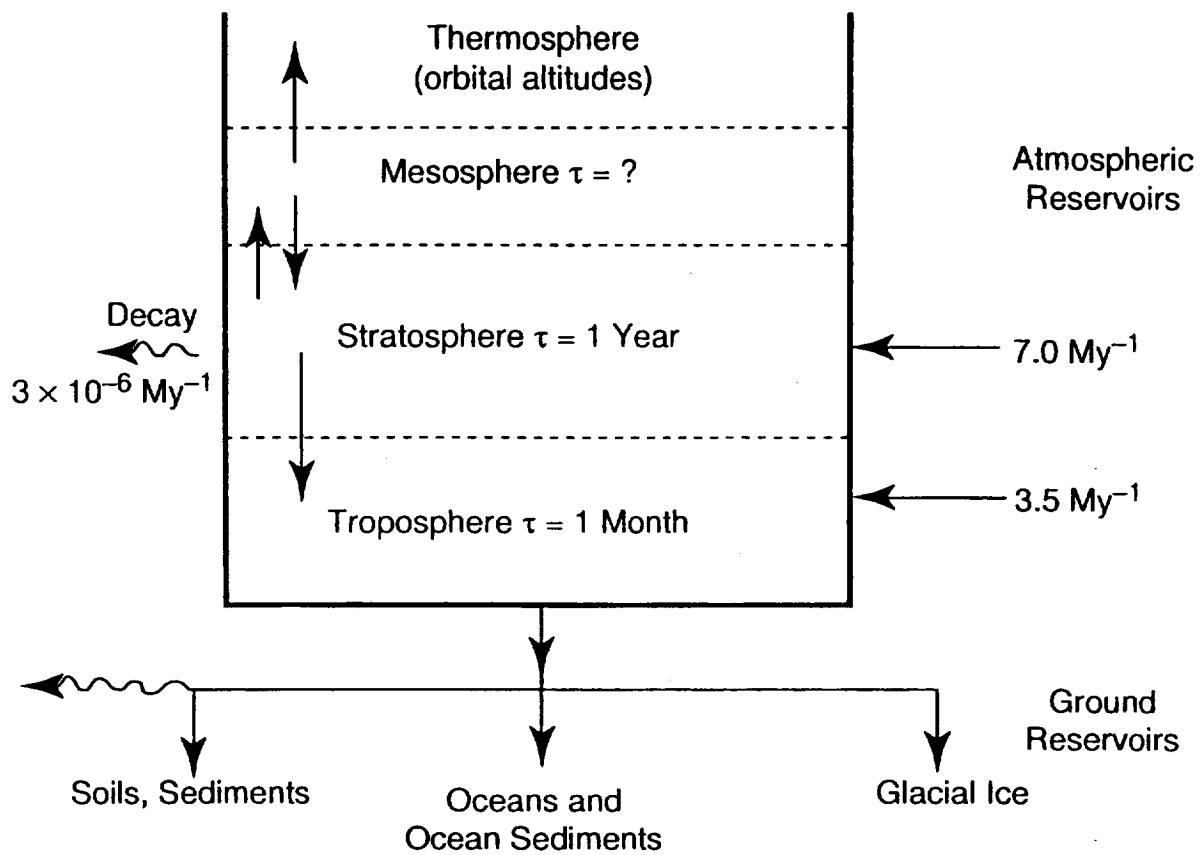


Fig 5